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Analysis of the Variability of Classified and Unclassified Radiological Source term Inventories in the Rainier Mesa/Shoshone Mountain Area, Nevada Test Site

P. Zhao, M. Zavarin

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Letter Report

Analysis of the Variability of Classified and Unclassified Radiological Source Term Inventories in the Rainier Mesa/Shoshone Mountain Area, Nevada Test Site

Pihong Zhao and Mavrik Zavarin
Chemistry, Materials, Environmental and Life Sciences Directorate, Lawrence Livermore National Laboratory

It has been proposed that unclassified source terms used in RM/SM reactive transport modeling investigations should be based on yield-weighted source terms calculated using the RM/SM average source term from Bowen et al. (2001) and the unclassified announced yields reported in DOE/NV-209. This unclassified inventory is likely to be used in unclassified contaminant boundary calculations and is, thus, relevant to compare to the classified inventory. We have examined the classified radionuclide inventory produced by 73 underground nuclear tests conducted in the Rainier Mesa / Shoshone Mountain (RM/SM) area of the Nevada Test Site. Our goals were to (1) evaluate the variability in classified radiological source terms among the 73 tests and (2) compare that variability and inventory uncertainties to an average unclassified inventory (e.g. Bowen 2001).

To evaluate source term variability among the 73 tests, radiological inventories were compared on two relative scales: geometric mean and yield-weighted geometric mean. Furthermore, radiological inventories were either decay corrected to a common date (9/23/1992) or the time zero (t_0) of each test. Thus, a total of four data sets were produced. The date of 9/23/1992 was chosen based on the date of the last underground nuclear test at the Nevada Test Site. The geometric mean activity for each radionuclide was calculated as follows:

$$GM_{RN} = \sqrt[n]{\prod_i A_{RN,i}} \qquad (1)$$

where $A_{RN,i}$ is the activity (A) for a particular radionuclide (RN) for test i and n is the total number of tests. A yield-weighted geometric mean activity was calculated as follows:

$$GM_{RN,Y} = \sqrt[n]{\prod_i \frac{A_{RN,i}}{Y_i}} \qquad (2)$$

where Y_i is the classified yield of test i. The relative inventory for each radionuclide and each test was calculated by dividing the test-specific radionuclide activity (or yield-weighted activity) by the associated geometric mean activity:

$$\log Ratio_{RN,i} = \log\left(\frac{A_{RN,i}}{GM_{RN}}\right) = \log A_{RN,i} - \log GM_{RN} \qquad (3)$$

$$Standard\ Deviation\ of\ log(Ratio_{RN,,i}) = \sqrt{\frac{\sum(\log Ratio_{RN,i} - \overline{\log Ratio_{RN,i}})^2}{n-1}} \qquad (4)$$

or the yield-weighted geometric mean activity:

$$\log Ratio_{RN,Y,i} = \log(\frac{A_{RN,i}/Y_i}{GM_{RN,Y}}) \ = \log(\mathrm{A}_{RN,\ i}/\mathrm{Y}) - \log\mathrm{GM}_{RN,Y} \qquad (5)$$

$$Standard\ Deviation\ of\ log(Ratio_{RN,Y,i}) = \sqrt{\frac{\sum(\log Ratio_{RN,y,i} - \overline{\log Ratio_{RN,Y,i}})^2}{n-1}} \qquad (6)$$

As stated earlier, relative inventories and yield-weighted relative inventories were calculated at a single date (9/23/1992) and at the t_0 for each test. This analysis generated four datasets (1992, t_0, yield weighted1992, yield weighted t_0) which could be discussed in an unclassified format because classified test-specific source term information is not available from the relative scale employed here.

All calculations were performed on a total of 33 radionuclides. Some 9 radionuclides including 3 activation products ^{26}Al, ^{93m}Nb, ^{150}Eu, and 6 actinides ^{232}U, ^{233}U, ^{236}U, ^{237}Np, ^{243}Am and ^{244}Cm, were excluded due to data inadequacies. ^{40}K, as a natural radionuclide, was also excluded. The activities of the excluded radionuclides account for < 0.1% of the total inventory. The removal of these radionuclides allowed us to focus on those radionuclides that are more relevant to modeling. For radionuclides that included both natural and device sources (^{232}Th, ^{235}U, and ^{238}U), the combined natural + device activity was used in this analysis. The relative activity results (Figure 1) suggest that, at RM/SM, radionuclide activities for individual tests typically fall within a 5 order of magnitude range. This variability is substantially larger than the inventory uncertainties reported in Bowen et al. (1992) (Table 1) and may reflect differences in test yield, performance, and individual radionuclide, etc.

Table 1. Estimated classified inventory accuracies for various groups of radionuclides. From Bowen et al. (2001).

	% Error	Order of magnitude
Fission Products	~10 to 30%	0.04-0.11
Unspent Fuel	20% or better	< 0.08
Fuel Activation Products	50% or better	< 0.18
Residual Tritium	300% or better	< 0.48
Activation Products	A factor of 10	1

Importantly, it is clear that the variability of the relative activity in fission product source term is dramatically reduced when weighted by yield. Figure 2 shows yield-weighted radionuclide activities relative to yield-weighed geometric mean decay corrected to September 23, 1992. Similar plots were obtained for yield weighted relative activities at t_0, however, the plots are not shown in this report. It should not be surprising given that the production of fission products should be strongly correlated to the quantity of fissioned device fuel. Furthermore, uncertainty in the classified source terms is relatively small (~10 to 30%, as reported in Bowen et al., 2001) when compared to the range of test yields at RM/SM. Thus, the relationship between yield and fission product activity is clearly discernible. Variability in activation products reduces to a range of 4 orders of magnitude, much less decrease than the one in fission products. The range of tritium relative activity does not appear to decrease when weighted by yield. For actinides, the residual actinide activity may, in fact, be negatively correlated with yield since efficient and large detonations might result in very little residual fuel. Activation product variability decreases somewhat when weighted by yield. However, activation product source terms are inherently uncertain; Bowen et al. (2001) estimated the activation product uncertainty for most NTS tests to be a factor of ~10. Thus, the uncertainty in source terms will minimize any reductions in variability of yield-weighted activation product calculations.

To facilitate further interpretation of source term variability, we partitioned the 33 radionuclides into 4 groups. The four groups were based on their primary source, as defined in Bowen et al. (2001): Tritium, activation products, fission products, and actinides. The 4 groups are the following:

1. Tritium: ^{3}H
2. Activation Products: ^{14}C, ^{36}Cl, ^{39}Ar, ^{41}Ca, ^{59}Ni, ^{63}Ni, ^{94g}Nb, ^{152}Eu, ^{154}Eu and ^{166m}Ho, total 10 radionuclides.
3. Fission Products: ^{85g}Kr, ^{90}Sr, ^{93}Zr, ^{99}Tc, ^{107g}Pd, ^{113m}Cd, ^{121m}Sn, ^{126}Sn, ^{129}I, ^{135g}Cs, ^{137}Cs, and ^{151}Sm, total 12 radionuclides.
4. Actinides: ^{232}Th, ^{234}U, ^{235}U, ^{238}U, ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu and ^{241}Am, total 10 radionuclides.

Some radionuclides are products of both activation and fission. We assigned these radionuclides according to their primary source. A relative activity range and standard deviation was calculated for each of the 4 radionuclide groups (Table 2). Calculations were also performed on U12t tunnel (6 tests) and U12n tunnel (22 tests) tests to compare variability between two prominent tunnel complexes.

Table 2 summarizes the relative activity ranges and standard deviations for each radionuclide group for all RM/SM tests. Similar data sets were also calculated for U-12n tunnel tests, and U-12t tunnel tests. With a decrease in the number of tests in U-12n and U-12t tunnels, the relative activity ranges and the standard deviations decreased as well. However, the data are not shown in this report.

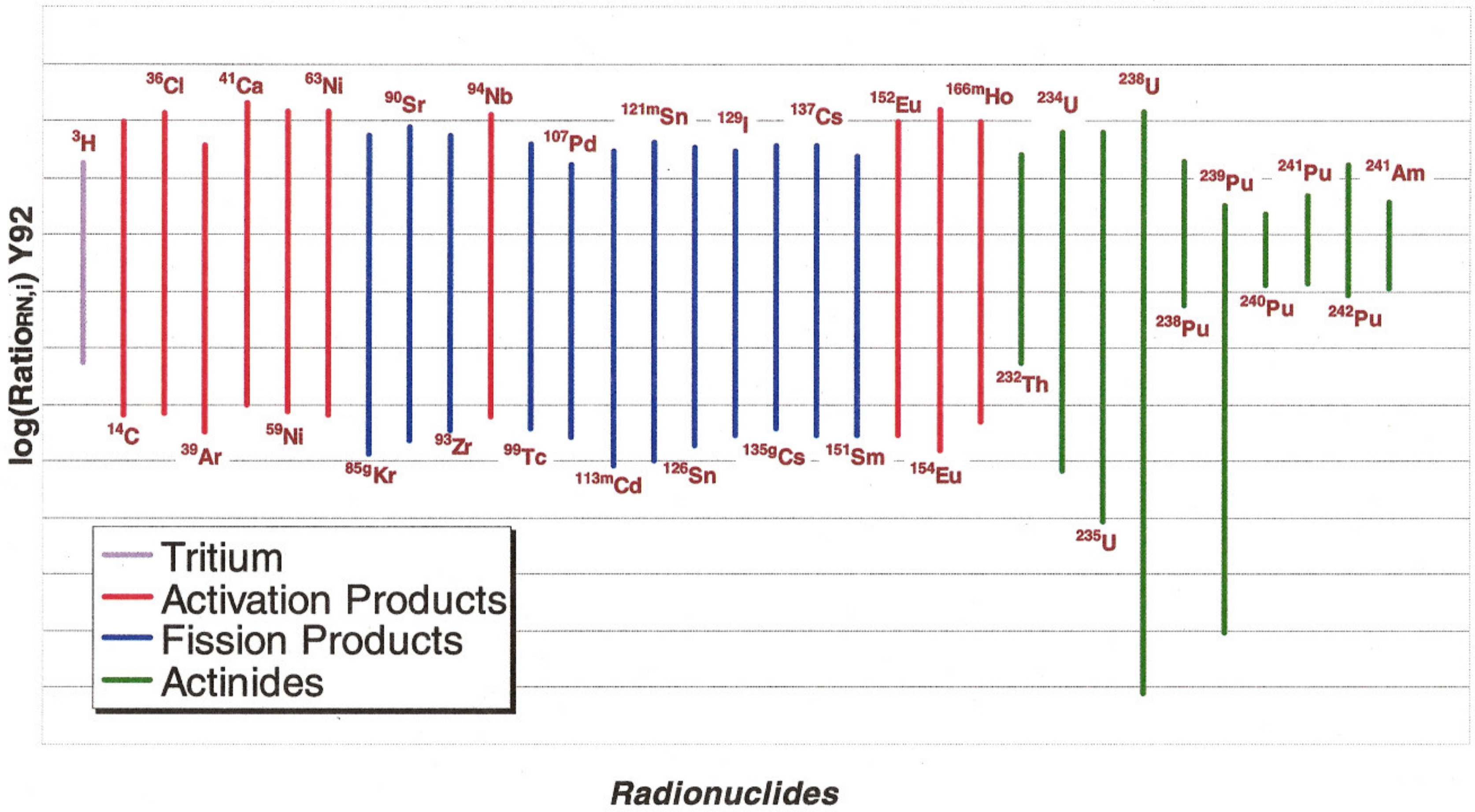

Figure 1. Ranges of radionuclide activities relative to geometric mean decay corrected to September 23, 1992.

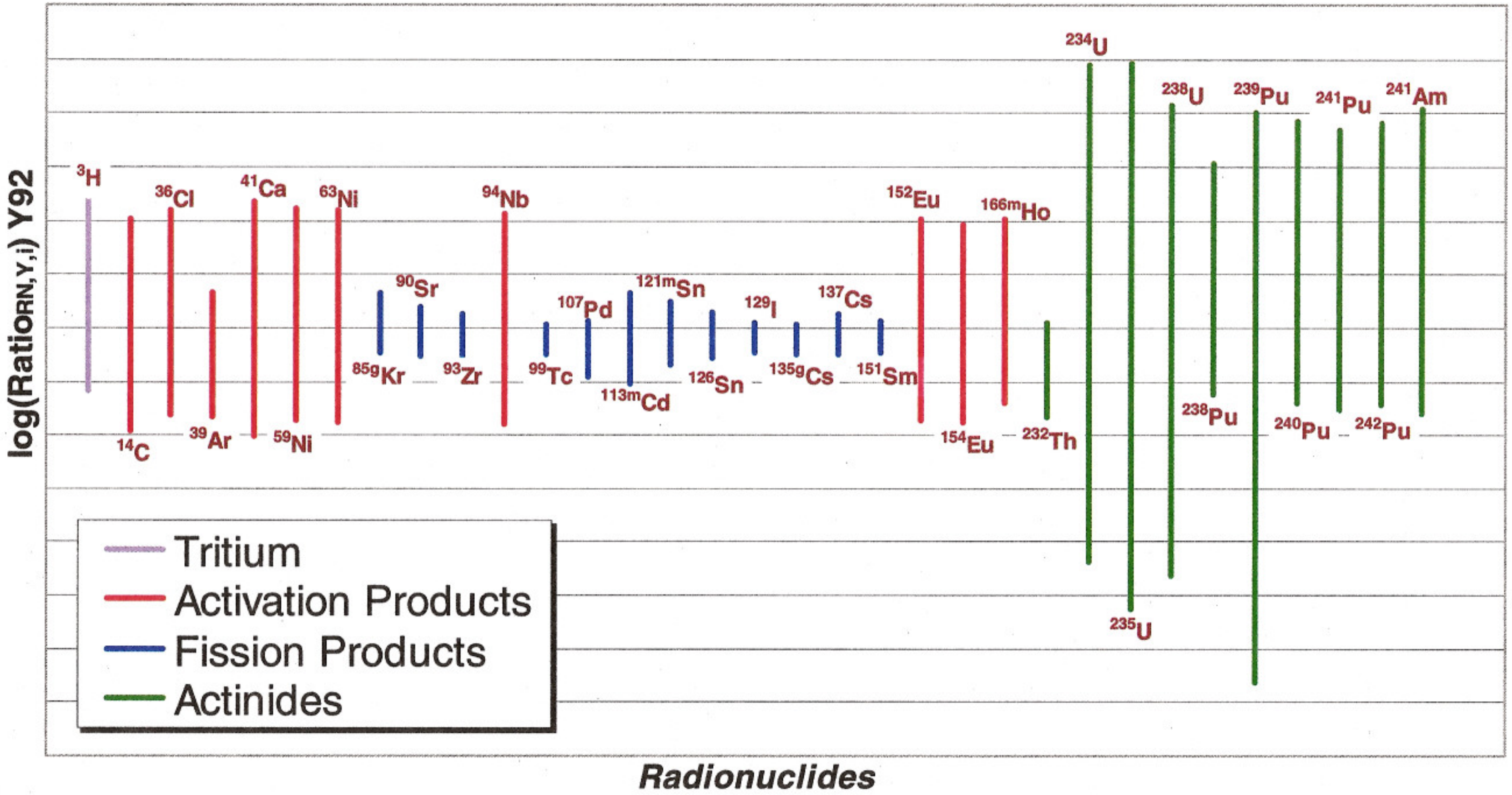

Figure 2. Ranges of yield-weighted radionuclide activities relative to yield-weighed geometric mean decay corrected to September 23, 1992.

Table 2. Range of Radionuclide Inventory at Rainier Mesa / Shoshone Mountain

		Range of Log(*Ratio*$_{RN,i}$) (Y92)*	Standard Deviation of log unit	Range of Log(Ratio$_{RN,Y,i}$) (Y92)*	Standard Deviation of log unit
Total RM Area	Tritium	3.52	0.74	3.88	0.74
73 tests	Activation Products	6.11	1.40	4.37	0.64
	Fission Products	5.96	1.27	1.72	0.21
	Actinides	10.25	0.82	11.56	1.23

units are order of magnitude

In general, yield weighting or decay-correction did not appreciably reduce the range of tritium activity. The variability is at a scale similar to the tritium inventory accuracy reported by Bowen et al. (2001) and, thus, may simply reflect source term uncertainty. Interestingly, the range of tritium activity in U-12t tunnel tests was substantially lower than U-12n tunnel tests. However, this may simply result from the small number of tests detonated in U-12t tunnel. Yield-weighting of activation products resulted in substantially narrower ranges, reducing the standard deviation by nearly half. The remaining variability can, most likely, be attributed to the large uncertainty in source term (Table 1). Yield weighting the fission products had an even more dramatic effect; standard deviations decreased to less than 0.2 log units in most cases (see Figure 2). The activities of fission products are clearly proportional to the reported test yields. Again, the remaining variability can, most likely, be attributed to source term uncertainties (Table 1). The relative activity ranges of actinides nearly doubled from time zero to the reference date of 9/23/1992 for tests in RM/SM and U-12n tunnels. It remained the same for tests in U-12t tunnels (6 tests only). More importantly, yield weighting of actinides substantially increased the actinide activity range and standard deviations. Thus, actinide activities are not proportional to test yield. In fact, tests with higher yields may, in some cases, have smaller actinide residual source terms because they effectively burn their actinide fuels. The exception in the actinide group is ^{232}Th, whose variability decreased after yield-weighting, most likely because the ^{232}Th inventory is predominantly of natural origin.

As general guidelines, yield-weighting of fission product radionuclides addresses a large proportion of the source term variability in RM/SM tests. Yield-weighting of activation product radionuclides also addresses some of the variability. The remaining variability can, most likely, be attributed to uncertainties in the classified inventories, as defined in Table 1. Importantly, yield-weighting addresses very little or none of the variability in tritium and actinide source terms. Thus, yield weighting is not an appropriate means for addressing actinide and tritium source term variability at RM/SM. Importantly, the yields used here were classified. Conducting this analysis using announced unclassified yields (i.e. DOE/NV-209) would substantially increase the variability in all yield-weighted calculations (see discussion below).

Histograms of radionuclide relative activity on reference date of 9/23/1992 are plotted in Figures 3 and 4. In most cases, the histograms show that activity distributions are asymmetric, especially, for groups of activation and fission products. The comparison between Figure 3 and Figure 4 indicates that the yield-weighting both activation and fission products substantially narrows the range of activity. This suggests that yield weighting addresses most of the source term variability for fission products and activation products. For tritium and actinides, yield-weighting increases variability; yield-weighting these radionuclides is not recommended. The similar conclusions can be drawn from analysis of U-12n tunnel and U-12t tunnel tests. However, the histograms for any tunnel area are not shown in this report.

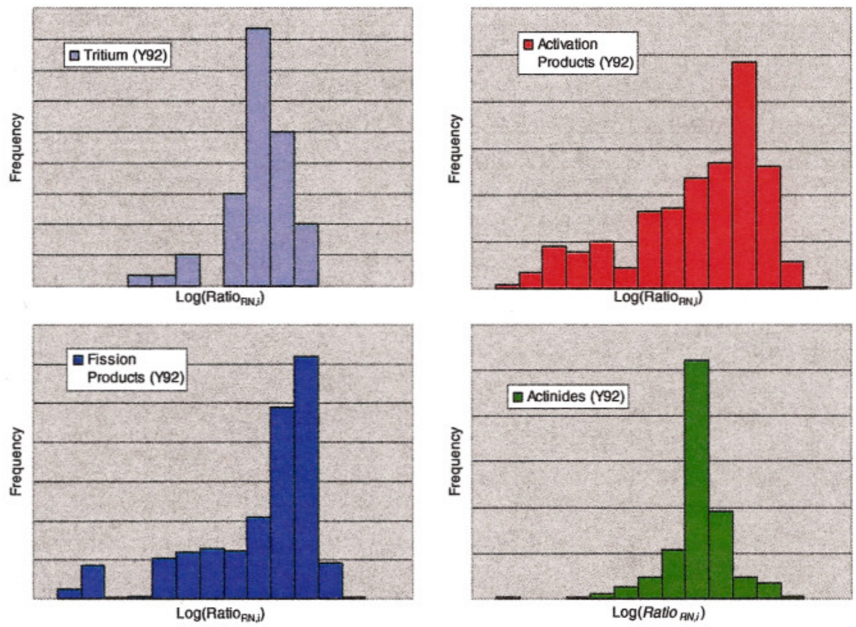

Figure 3. Histograms of radionuclide activities relative to their geometric means decay corrected to 9/23/1992.

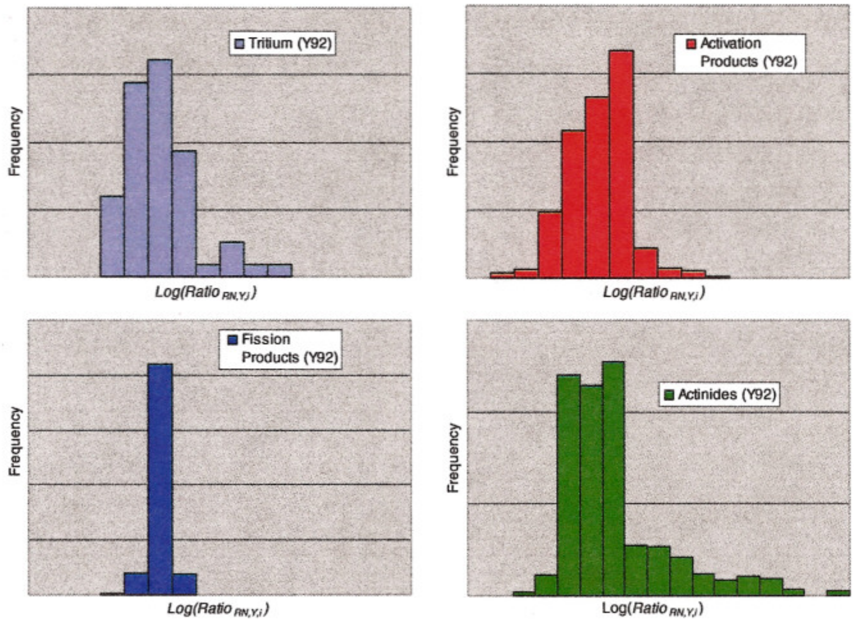

Figure 4. Histograms of yield-weighted radionuclide activities relative to their yield-weighted geometric means decay corrected to 9/23/1992.

It has been proposed that unclassified source terms used in RM/SM reactive transport modeling investigations should be based on yield-weighted source terms calculated using the RM/SM average source term from Bowen et al. (2001) and the unclassified announced yields reported in DOE/NV-209. This unclassified inventory is likely to be used in unclassified contaminant boundary calculations and is, thus, relevant to compare to the classified inventory. To calculate the unclassified yield-weighted inventory for each test and each of the 33 radionuclides, the following assumption were made:

1. Many of the announced yields reported in DOE/NV-209 are, in fact, ranges. The maximum yield of the range was used here.
2. A yield of 1 ton (the lowest yield reported in RM/SM) was assigned to events Saturn and Mercury. Event Saturn was reported zero yield in DOE/NV-209, and event Mercury, for which a numerical value was not reported in DOE/NV-209.

The unclassified yield-weighted inventory of all RM/SM tests and all 33 radionuclides examined previously was compared to the classified inventory. Table 3 summarizes 95% confidence intervals for correlation of unclassified yield-weighted activities and their classified counterpart for tests in RM/SM area (in log unit). The behavior of ^{59}Ni was substantially different from other activation products; it was eliminated from this analysis. Importantly, most classified radionuclide source terms fall within an order of magnitude of the unclassified yield-weighted average. This is encouraging that it suggests that unclassified contaminant boundary calculations are not likely to differ dramatically from their classified counterparts.

Table 3. 95% Confidence Intervals for correlation of unclassified yield-weighted activities and classified activities for tests in RM/SM area (log unit).

# of tests	Tritium	Activation Products (exclude ^{59}Ni)	Fission Products	Actinides
73 tests	2.02	2.34	1.77	2.68

The confidence intervals for correlations between unclassified yield-weighted inventories and classified inventories are plotted for tritium, activation products, fission products, and actinides in Figures 5 through 8. The lines represent 68% (red) and 95% (blue) confidence intervals (CI), respectively, for each radionuclide group. The unclassified yield-weighted inventory is calculated for each test and each radionuclide using the average Rainier Mesa/Shoshone Mountain source term (Bowen et al., 2001) and the maximum announced yield (DOE/NV-209):

$$A_{U,RN,i} = \frac{A_{U,RN,T}}{Y_{U,T}} Y_{U,i} \qquad (7)$$

Where $A_{U,RN,T}$ is the total Rainier Mesa/Shoshone Mountain activity for a radionuclide (Bowen et al., 2001), $Y_{U,T}$ is the total unclassified yield based on maximum announced yields (DOE/NV-209), and $Y_{U,i}$ is the unclassified maximum announced yield for test i.

The confidence intervals are calculated from the log-scale differences between classified inventories and unclassified yield weighted inventories for all ten tests ($\log A_{RN,i} - \log A_{U,RN,i}$). Based on this analysis, it is apparent that most unclassified radionuclide inventories fall within an order of magnitude of their classified counterparts (Table 3). Not surprisingly, the confidence interval for fission products is the smallest of all radionuclide groups. As shown in the previous analyses, actinide source term variability in RM/SM tests is not a function of test yield. Thus, the yield-weighted unclassified source term does not correlate strongly with the classified inventory (Figure 8). Nevertheless, 73% of all actinide test inventories fall within one order of magnitude of their respective classified counterparts. Fission product inventories correlate very strongly with yield (Figure 7). Thus, 84% of all fission product inventories fall within one order of magnitude of their respective classified counterparts. Tritium and activation product radionuclide source terms correlate with yield (Figure 5 and 6). However, there is substantial uncertainty in the classified estimates of both tritium and activation products. Thus, the correlations are weaker than for the fission products.

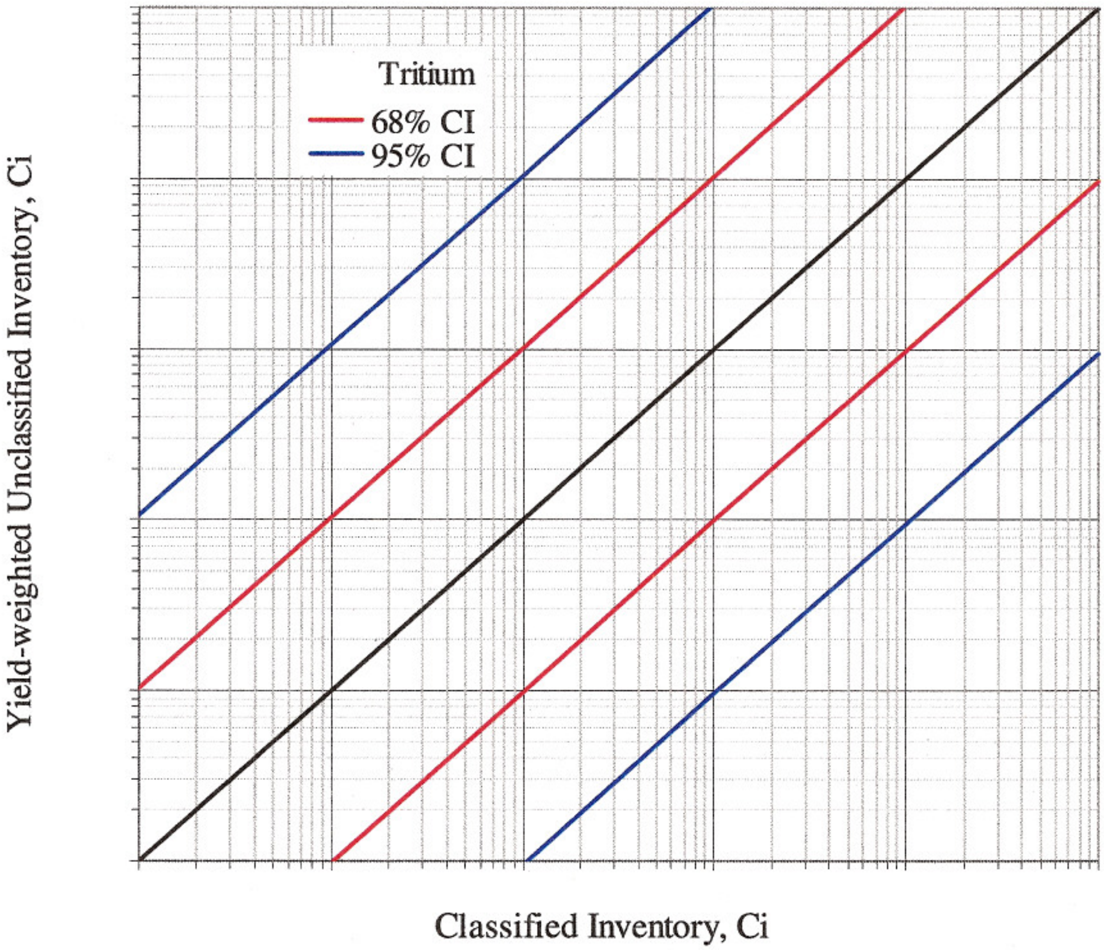

Figure 5. Tritium source term correlation between classified inventories and yield-weighted unclassified inventories for tests detonated in Rainier Mesa/Shoshone Mountain. Red lines and blue lines represent 68% and 95% confidence intervals (CI), respectively.

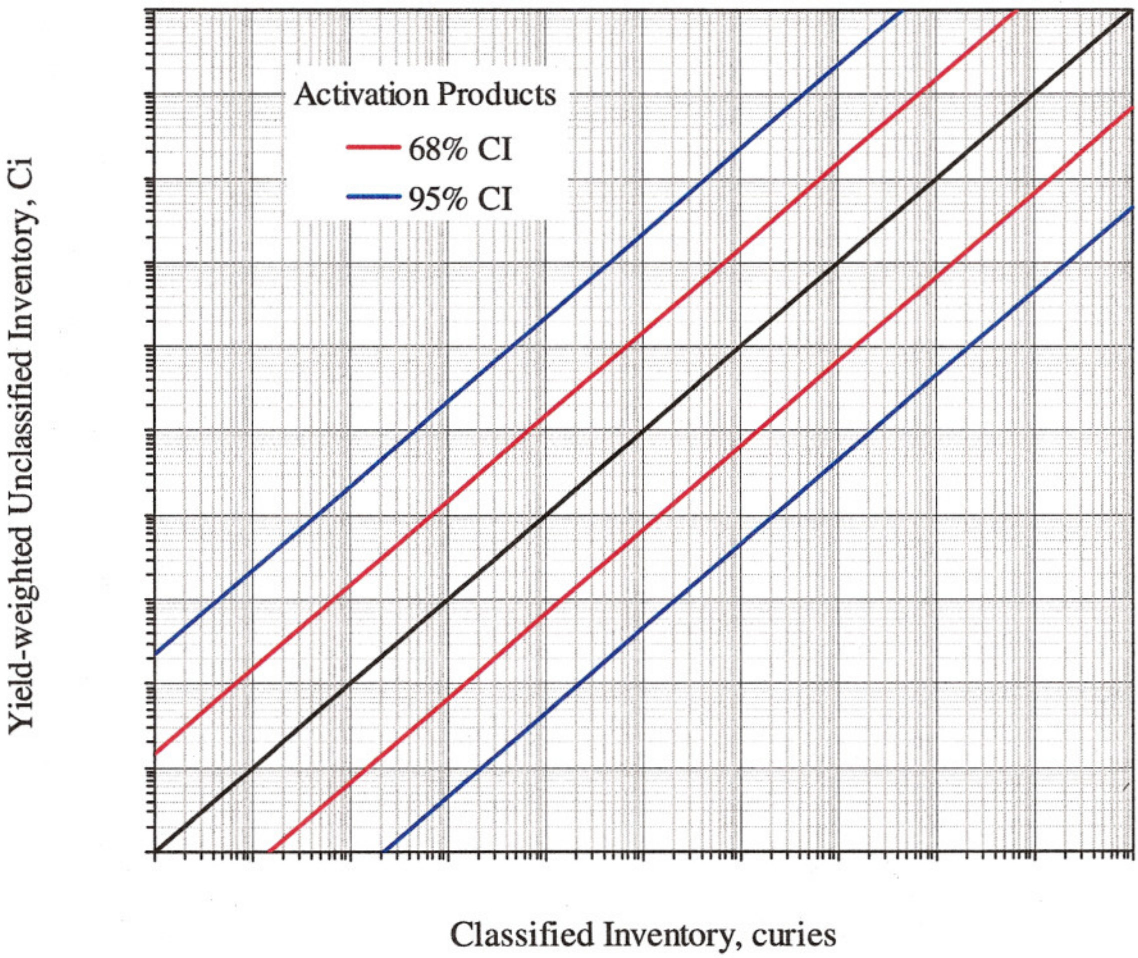

Figure 6. Activation product source terms correlation between classified inventories and yield-weighted unclassified inventories for tests detonated in Rainier Mesa/Shoshone Mountain. Red lines and blue lines represent 68% and 95% confidence intervals, respectively.

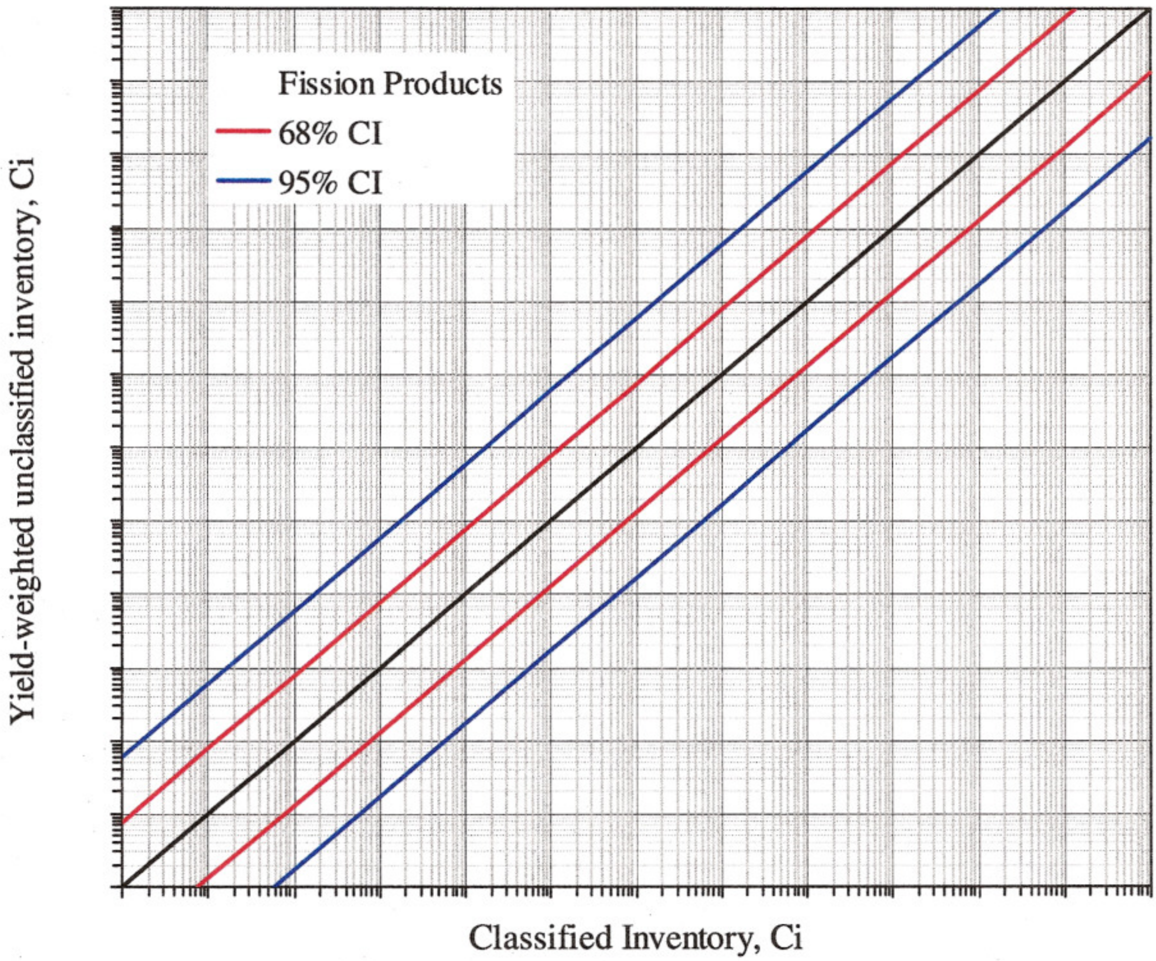

Figure 7. Fission product source term correlation between classified inventories and yield-weighted unclassified inventories for tests detonated in Rainier Mesa/Shoshone Mountain. Red lines and blue lines represent 68% and 95% confidence intervals, respectively.

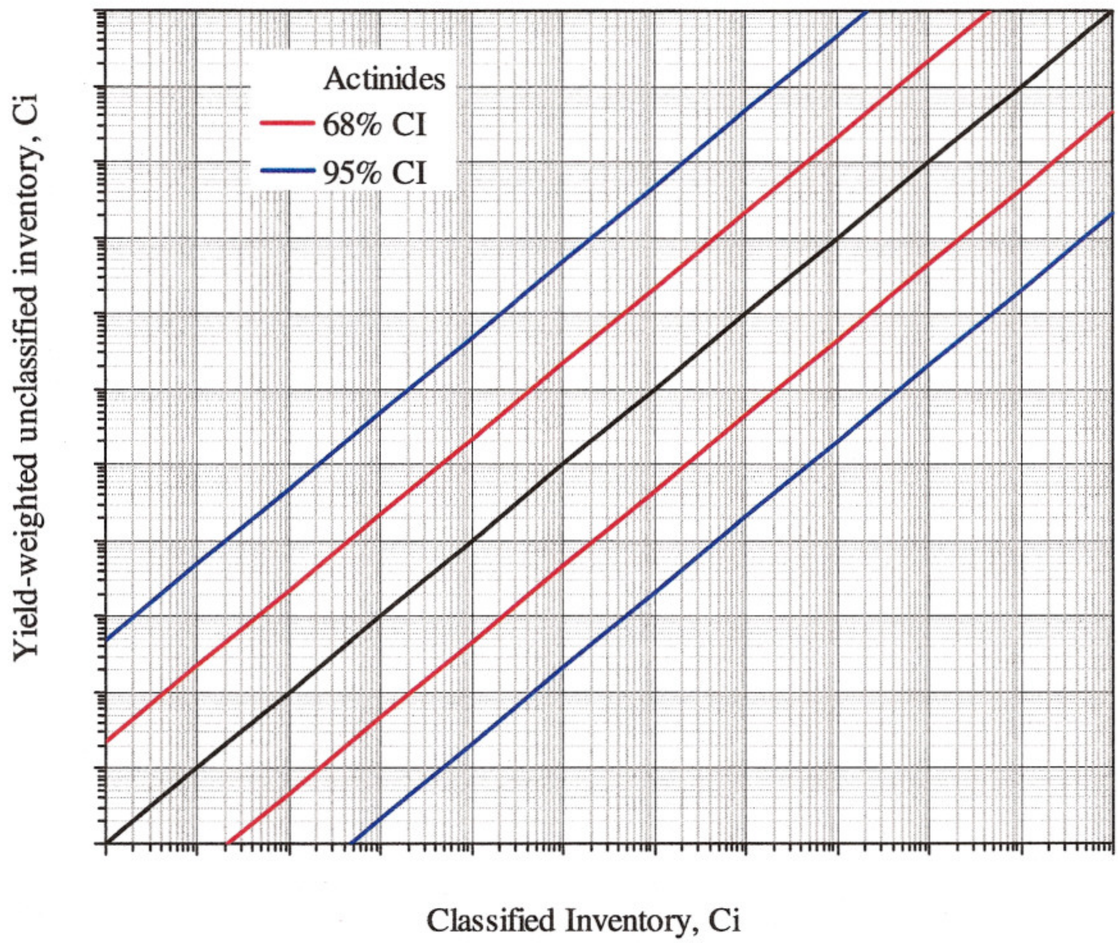

Figure 8. Actinide source terms correlation between classified inventories and yield-weighted unclassified inventories for tests detonated in Rainier Mesa/Shoshone Mountain. Red lines and blue lines represent 68% and 95% confidence intervals, respectively.

CONCLUSIONS

Based on our analysis of yield-weighted classified inventories, we find that much of the variability in fission and activation product inventories can be attributed to test yield. The remaining variability can be attributed to the uncertainty in classified source term estimates (i.e. Table 1). However, actinide fuel variability cannot be attributed to yield. This is likely because efficient high-yield tests would tend to burn up most of their fuel, resulting in low actinide concentrations. Conversely, tests that did not perform to specifications may have low yields and significant amounts of unburned fuel. Tritium source terms do not correlate very strongly with yield, in part due the large uncertainties associated with its classified inventory and its different uses and sources on various tests.

When comparing classified source terms to the unclassified yield-weighted source terms calculated using information from Bowen et al. (2001) and DOE/NV-209, we find that a majority of unclassified radionuclide inventories fall within one order of magnitude of the classified inventory. This is encouraging in that it suggests that unclassified contaminant boundary calculations are not likely to differ dramatically from their classified counterparts.

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